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Thermal Conductivity of the Pyrochlore Superconductor KOs_2O_6: Strong Electron Correlations and Fully Gapped Superconductivity

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To elucidate the nature of the superconducting ground state of the geometrically frustrated pyrochlore KOs_2O_6 ($T_c = 9.6$ K), the thermal conductivity was measured down to low temperatures ($\sim T_c/100$). We found that the quasiparticle mean free path is strikingly enhanced below a transition at $T_p = 7.8$ K, indicating enormous electron inelastic scattering in the normal state. In magnetic fields, the conduction at $T \to 0$ K is nearly constant up to $\sim 0.4H_{c2}$, in contrast with the rapid growth expected for superconductors with an anisotropic gap. This unambiguously indicates a fully gapped superconductivity, in contrast to the previous studies. These results highlight that KOs_2O_6 is unique among superconductors with strong electron correlations.

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Geometrically frustrated systems have recently become the subject of intense theoretical and experimental study. A fundamental question for such systems is the nature of the ground state. For itinerant electron systems, it has been argued that geometrically frustrated lattice gives rise to exotic phenomena including heavy fermion states [1], metal-insulator transitions [2], and the anomalous Hall effect [3]. The pyrochlore system is an ideal system for such studies, since the network of the relevant metal atoms consists of corner sharing tetrahedra. Very recently, superconductivity has been discovered in the β-pyrochlore oxides $R$$Os_2O_6$ (R = Cs, Rb, and K) [4, 5, 6, 7, 8]. These compounds have attracted great interest because the geometrical frustration inherent to the crystal structure may give rise to an exotic superconducting state.

$CsOs_2O_6$ (T_c = 3.3 K) and $RbOs_2O_6$ (T_c = 6.3 K) show rather usual behavior. In both compounds, T^2-dependent resistivity ρ is observed and the upper critical fields H_{c2} are below the Pauli limit. A jump ΔC in the specific heat at T_c implies superconductivity in the intermediate coupling regime. Several experiments indicate the conventional BCS s-wave superconductivity [9, 10, 11, 12, 13].

In sharp contrast, the behavior of KOs_2O_6 with the highest T_c (= 9.6 K) appears to be highly unusual. KOs_2O_6 exhibits an extremely low-energy large rattling motion of the K ions in an oversized cage forming a three dimensional skeleton [14]. In the normal state, the resistivity exhibits a strong convex T-dependence from just above T_c extending to room temperature, indicating anomalous electron scattering [6]. Specific heat measurement has revealed an unusually large mass enhancement with a Sommerfeld coefficient of γ =70-110 mJ/K^{2}mol, which is strongly enhanced from the band calculation value of 9.8 mJ/K^{2}mol [15, 16]. In addition, strong coupling superconductivity has been suggested based on the large ΔC [12, 15]. With decreasing T, H_{c2} increases linearly even below 1 K, showing no saturation [17]. Moreover, $H_{c2} \simeq 32$ T at $T \to 0$, exceeding the apparent Pauli limited value ~ 18 T. Measurements of λ by μSR [9] and NMR relaxation rate T_1^{-1} [18] suggest a very anisotropic gap function, although low-temperature measurements below 2 K are lacking. Very recently, a first-order phase transition, which occurs at $T_p \sim 7.5$ K below T_c, has been reported [15]. This transition is insensitive to magnetic field and it is suggested that it can be associated with the rattling of the K atoms, though the details are unknown.

Thus a major outstanding question is how geometrical frustration and the rattling motion affect the superconducting state in KOs_2O_6, including the microscopic pairing mechanism. To clarify this issue, a detailed study of the low-energy quasiparticle (QP) properties is of primary importance. We here report the QP and phonon transport probed by the thermal conductivity. We observed anomalous QP dynamics in the superconducting state suggesting inherent strong electron correlations that usually prefer anisotropic superconductivity. However, we found strong evidence for an isotropic gap. These contrasting results highlight the distinct superconducting state in KOs_2O_6.

Thermal conductivity κ was measured by a standard steady-state method in single crystals of KOs_2O_6 with cubic structure grown by the technique described in [15]. The contact resistance was less than 0.1 Ω at low temperatures. The magnetic field H was applied parallel to the current direction to minimize the vortex motion. As shown in the inset of Fig. 1, anomalous convex resistivity $\rho(T)$ from just above T_c up to 300 K is observed [6]. The main panel of Fig. 1 depicts the T-dependence of κ, together with C/T in zero field. Both κ and C/T exhibit distinct anomalies at T_c and T_p. Upon entering the superconducting state, $\kappa(T)$ changes its slope with a cusp at T_c and decreases with decreasing T, followed by a kink near T_p. Below T_p, $\kappa(T)$ increases gradually, peaks at $\sim$ 7 K, and then decreases rapidly down to $\sim$3 K. Below $\sim$ 3 K, it decreases gradually. The Lorentz number $L = \kappa\rho/T = 1.1L_0$ at T_c is close to the Sommerfeld value $L_0 = 2.44 \times 10^{-8}$ ΩW/K^2, indicating substantial electronic contribution in the heat conduction near T_c.

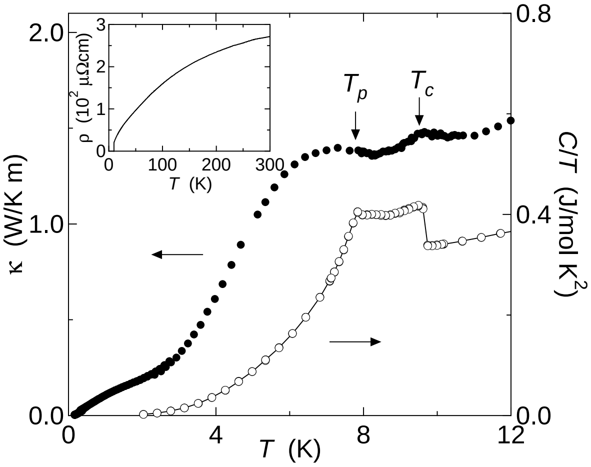

FIG. 1: Zero-field thermal conductivity and specific heat in a KOs_2O_6 crystal ($0.3 \times 0.37 \times 0.88$ cm^3). Inset: Temperature dependence of the resistivity.

In Fig. 2(a), the T-dependence of $\kappa(T)/T$ for several magnetic fields is plotted. In zero field, κ/T displays a steep increase below T_p and exhibits a pronounced maximum at $\sim$ 6 K. At very low temperatures, $\kappa(T)/T$ decreases rapidly with decreasing T after showing a second maximum at $\sim$ 0.8 K. The peak structure in κ/T below $T_c(H)$ disappears above 1 T, indicating that the enhancement is readily suppressed by a magnetic field.

We first discuss the origin of the double peak structure in κ/T below T_p in zero field. The maximum in κ/T at $\sim$ 0.8 K is attributed to the phonon peak. A peak appears when the phonon conduction grows rapidly at very low temperatures and is limited by the sample boundaries. Such a phonon peak well below T_c has been reported for Nb and $LuNi_2B_2C$ [19, 20]. Below $\sim$ 250 mK, the phonon contribution $\kappa_{ph} = bT^3$ can be separated from the electronic one $\kappa_e = aT$ by a fit $\kappa/T = a + bT^2$ in Fig. 2(b). From the coefficient b, we obtain a reasonable value of the acoustic phonon velocity $v_s \simeq 4700$ m/s [20].

The other peak in κ/T at $\sim$ 6 K is most likely to be of electronic origin, because (1) the appearance of phonon peaks twice below T_c is highly unlikely, and (2) the electronic contribution is substantial near T_c. The electronic heat conduction is described by $\kappa_e/T \sim N(0)v_F^2\tau_e$, where $N(0)$, v_F, and τ_e are the QP density of states (DOS) at the Fermi level, Fermi velocity, and QP scattering time, respectively. Therefore the enhancement of κ_e/T is a strong indication that a striking enhancement of τ_e occurs overcoming the reduction of $N(0)$ in the superconducting state of KOs_2O_6. Recent microwave measurements also show the enhancement of the QP conductivity [21] consistent with our observation.

We here examine the T-dependence of τ_e quantitatively below T_c, assuming $\kappa \simeq \kappa_e$. In the temperature range $T_p < T < T_c$, κ/T decreases from that extrapolated above T_c, indicating that the thermal conductivity is mainly determined by the change of $N(0)$. We note

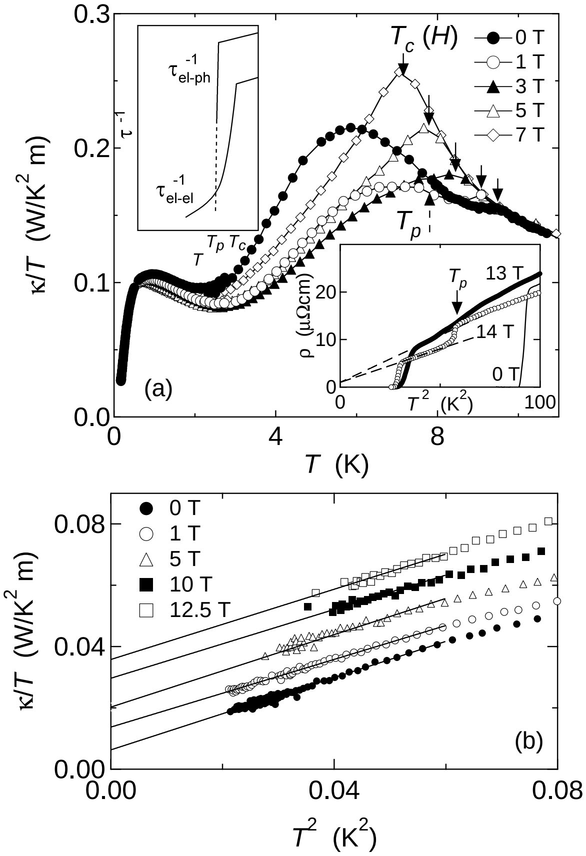

FIG. 2: (a) T-dependence of κ/T in various magnetic fields. The superconducting transition temperatures $T_c(H)$ are shown by arrows. T_p (dashed arrow) is independent of H. Upper inset: schematic figure of T-dependence of $\tau_{\text{el-ph}}^{-1}$ and $\tau_{\text{el-el}}^{-1}$. Lower inset: ρ vs T^2 at 0 and 13 T (solid circles). Data for a different crystal at 14 T are also plotted (open circles). The dashed lines represent $\rho(T) = \rho_0 + AT^2$. (b) κ/T vs T^2 at low temperatures. The solid lines are linear fits.

that $N(0)$ is little affected by the phase transition at T_p, since H_{c2} changes only slightly and no discernible change of the penetration depth is observed [9, 15]. Therefore the enhancement of τ_e below T_p is responsible for the increase of κ/T. While $\kappa/T(0.6T_c)$ is enhanced nearly 1.5 times compared with $\kappa/T(T_c)$, $N(0)$ at $T = 0.6\ T_c$ is reduced to $\sim 1/4$ of $N(0)$ at T_c according to μSR measurements [9]. Hence τ_e is enhanced by 6 times at 0.6 T_c, indicating a remarkable enhancement in the superconducting state.

A rapid increase of τ_e indicates the presence of strong inelastic scattering originating from an interaction that has developed a gap [22]. We then expect two sources of inelastic scattering rate in KOs_2O_6, electron-phonon $\tau_{\text{el-ph}}^{-1}$ and electron-electron scattering rate $\tau_{\text{el-el}}^{-1}$, so that τ_e can be expressed as $\tau_e^{-1} = \tau_{\text{imp}}^{-1} + \tau_{\text{el-el}}^{-1} + \tau_{\text{el-ph}}^{-1}$, where τ_{imp}^{-1} is the impurity scattering rate. From the convex $\rho(T)$ and the large rattling motion of the K ions, we speculate that above T_p, the electron-phonon scattering

dominates, $\tau_{\text{el-ph}}^{-1} \gg \tau_{\text{imp}}^{-1}, \tau_{\text{el-el}}^{-1}$. Since the enhancement of phonon conductivity occurs at very low temperatures, as evidenced by the peak at $\sim T_c/10$, the increase of $\tau_{\text{el-ph}}$ just below T_c is expected to be small. This scenario is consistent with the slight change of κ/T just below T_c.

Below T_p, the scattering mechanism changes dramatically. In the lower inset of Fig. 2(a), we show $\rho(T)$ in two crystals at high fields where $T_c(H) < T_p$. The normal-state ρ well below T_p exhibits the Fermi-liquid behavior $\rho_0 + AT^2$ with $\rho_0 = 1.0$ $\mu\Omega$ cm and $A = 0.14$-0.20 $\mu\Omega$ cm/K^2. This, together with the large $\gamma \sim 100$ mJ/K^{2}mol [15, 16], follows the Kadowaki-Woods relation $A = a_{\text{KW}}\gamma^2$ with a_{KW} close to the universal value of 10^{-5} $\mu\Omega$ cm(K mol/mJ)^2 as shown in Fig. 3 [23]. This indicates that strongly correlated electrons with large mass are responsible for the T^2-dependence. Therefore it is natural to consider that below T_p electron-phonon scattering suddenly diminishes and QP transport is dominated by electron-electron scattering, as schematically shown in the upper inset of Fig. 2(a); the enhancement of κ/T stems from the enhancement of $\tau_{\text{el-el}}$. These results are also important for understanding the nature of the transition at T_p, namely our results support the proposed rattling transition because the freezing of the K ion motion should strongly influence the phonon spectrum.

To our knowledge, such enhancement of κ_e in the superconducting state has been reported only in very clean high-T_c cuprates [22] and heavy-fermion CeCoIn$_5$ [24]. We also note that the magnitude of the enhancement of τ_e observed in KOs$_2$O$_6$ is comparable to that in these systems, in which anisotropic d-wave pairing states are realized with strong electron correlations [25, 26].

Having established the evidence for strong electronic correlations, the next important issue is the paring symmetry. Owing to recent progress in understanding the thermal conductivity, it is well established that there is a fundamental difference in the heat transport between isotropic and anisotropic superconductors. We first discuss the gap function in terms of zero-field thermal conductivity at very low temperatures displayed in Fig. 2(b). As discussed above, we observe $\kappa/T = a + bT^2$ and the electronic contribution can be evaluated from the residual linear term $a = \kappa/T(T \to 0) \simeq 7 \times 10^{-3}$ W/K^{2}m. In unconventional superconductors with a line node, the universal residual linear term κ_{00}/T is present, which is independent of impurity concentration [27]. The question arises whether the finite a-term indicates a line node. To check this, we compare a with the linear term in the normal state κ_n/T. The Wiedemann-Franz law and $\rho_0 = 1.0$ $\mu\Omega$cm yield $\kappa_n/T =$ 2.44 W/K^{2}m, a value larger than a by a factor of more than 300. The residual linear term expected for a line node is given as $\frac{\kappa_{00}}{T} = \left(\frac{2}{\pi}\frac{\hbar}{\Delta\tau_{\text{imp}}}\right)\frac{\kappa_n}{T}$, where Δ is the superconducting gap. By using $\tau_{\text{imp}} = \mu_0\lambda^2/\rho_0$ with $\lambda = 270$ nm, and $\Delta = 1.9k_{\text{B}}T_c$ [16], κ_{00}/T is estimated to be ~ 0.07 W/K^{2}m. This value is an order of magnitude larger than a, indicating that low-temperature κ in zero field is totally incompatible with the line node. The origin of the small residual linear term may be due to a non-superconducting metallic regime in the crystal.

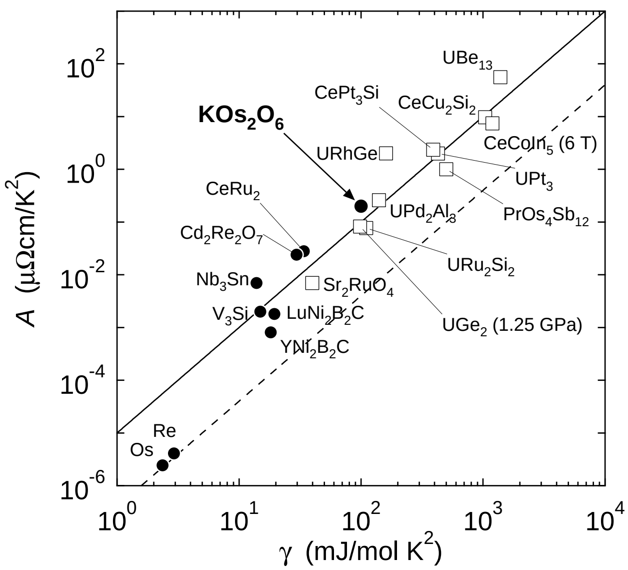

FIG. 3: Coefficient A vs Sommerfeld constant γ (Kadowaki-Woods plot [23]) for various superconductors. Solid circles are for superconductors believed to have isotropic gap (or anisotropic s-wave symmetry). Open squares for anisotropic gap with nodes. The lines represent $A = a_{\text{KW}}\gamma^2$ with $a_{\text{KW}} = 10^{-5}$ (solid) and 4×10^{-7} $\mu\Omega$ cm(K mol/mJ)^2 (dashed).

Strong evidence for fully gapped superconductivity is provided by the H-dependence of κ. Figure 4 depicts the H-dependence of κ/κ_n in the low-temperature limit. For comparison, $\kappa(H)/\kappa_n$ for several superconductors, s-wave Nb, MgB$_2$ with two gaps [28], and anisotropic UPt$_3$, LuNi$_2$B$_2$C [29], and CePt$_3$Si (line node) [30], are also plotted. We immediately notice that the H-dependence of $\kappa(H)/\kappa_n$ in KOs$_2$O$_6$ stays nearly constant up to $\sim 0.4H_{c2}$ and is very close to that of Nb, in dramatic contrast with those in anisotropic superconductors. In the superconducting state, the thermal transport is governed by the delocalized QPs, which extend over the whole crystal. In s-wave superconductors the only QP states present at $T \ll T_c$ are those associated with vortices. When the vortices are far apart, these states are bound to the vortex core and are therefore localized and unable to transport heat; the conduction shows an exponential behavior with very slow growth with H at $H \ll H_{c2}$, as observed in Nb. In sharp contrast, the heat transport in nodal superconductors is dominated by contributions from delocalized QP states. The most remarkable effect on the thermal transport is the Doppler shift of the energy of QPs [31, 32]. In the presence of line nodes where the DOS has a linear energy dependence ($N(E) \propto E$), $N(H)$ increases in proportion to $\sqrt{H}$. Therefore, the thermal conductivity in superconductors with large anisotropy grows rapidly at low field,

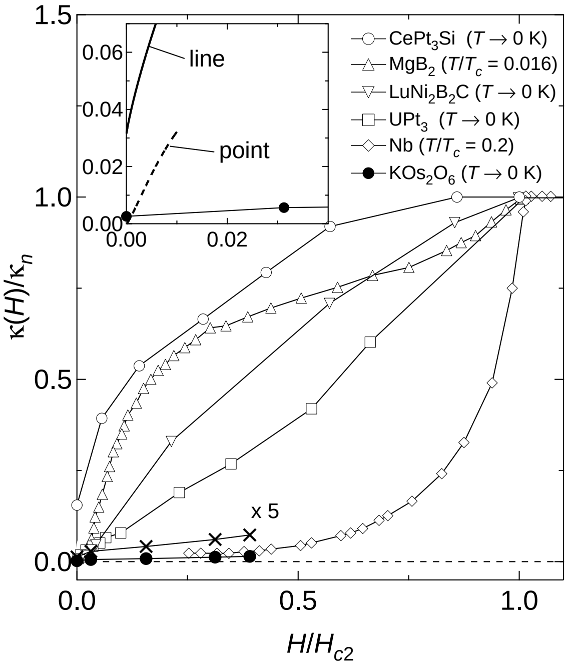

FIG. 4: H-dependence of κ/κ_n, as a function of H/H_{c2}. For comparison, data for the s-wave Nb, MgB_2 with two gaps, anisotropic $CePt_3Si$ (line node), $LuNi_2B_2C$ (anisotropic s), and UPt_3 are shown. Crosses indicate values for $\rho_0 = 5\ \mu\Omega$cm. Inset: H-dependence of κ at low fields. The solid and dashed lines represent the data for line and point nodes, respectively.

as shown in UPt_3, $LuNi_2B_2C$, and $CePt_3Si$. In the inset of Fig. 4 is plotted $\kappa(H)/\kappa_n$ at low fields for gap functions with line [31] and linear point [32] nodes for unitary limit calculated using the same parameters as adopted in the analysis of the residual κ/T. It is clear that $\kappa(H)/\kappa_n$ of KOs_2O_6 shows much slower growth than that of superconductors with point and line nodes. Thus no discernible delocalized QPs exist at least for $H < 0.4H_{c2}$ in KOs_2O_6. In case we have underestimated ρ_0 in the fit in the inset of Fig. 2, we also plot the case for ρ_0 5 times larger and find that the slope of κ/κ_n is still close to that of Nb. The marked insensitivity of κ to H, together with the tiny residual linear term, leads us to conclude that the gap function of KOs_2O_6 is fairly isotropic.

Strong electron correlations usually favor anisotropic superconductivity, since Coulomb repulsion prefers small probabilities in small pair distances. This is demonstrated in Fig. 3 by separating very anisotropic superconductors with nodes (p-, d-wave, etc.) from s-wave (and anisotropic s) superconductors by large $\gamma \gtrsim$ 30-40 mJ/mol K^2. Our results therefore highlight the unusual full-gap superconductivity with strong correlations characterized by large γ in KOs_2O_6.

In summary, we have investigated the superconducting state of KOs_2O_6 by thermal conductivity. In contrast to previous studies, the isotropic gap function is firmly indicated. We also found the striking enhancement of the QP scattering time in the superconducting state. The present results indicate that strong electron correlations coexist with a fully gapped superconductivity, which make KOs_2O_6 a quite unique system. How the unique superconductivity is related to geometrical frustration and rattling is an intriguing issue, which deserves further experimental and theoretical studies.

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